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Windsor [GB/ZA]; 16 Luipaards Avenue, Monument Park,
0181 Pretoria (ZA).

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(74) Agents: CLELLAND, Sandra, Luischen et al.; Spoor
and Fisher, P O Box 41312, 2024 Craighall (ZA).

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(71) Applicant (*for ZA only*): BALMORAL TECHNOLO-
GIES (PROPRIETARY) LIMITED [ZA/ZA]; Building
16, CSIR Campus, Meiring Naude Road, Scientia, 0002
Pretoria (ZA).

(71) Applicant (*for all designated States except US, ZA*):
WINDSOR TECHNOLOGIES LIMITED [BS/BS];
Sandringham House, 83 Shirley Street, Nassau (BS).

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(72) Inventor; and

(75) Inventor/Applicant (*for US only*): SYMONS, Michael,

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(57) Abstract: A method of making a finished product includes the steps of providing a feedstock comprising a polymer component comprising 5 to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; 0 to 95 parts by weight of polystyrene in particulate form; an extender; and optionally reinforcing fibres or particles; mixing the feedstock with a thermosetting resin selected from an unsaturated polyester resin in liquid form and a catalyst for the polyester resin and an epoxy resin in liquid form and a catalyst for the epoxy resin; and subjecting the product to suitable conditions of temperature and pressure to cause the polyethylene and/or the polyvinyl chloride and the polystyrene, if present, to melt and the thermosetting resin to set to form the finished product, e.g. a shutter board.

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METHOD OF MAKING A FINISHED PRODUCT**BACKGROUND OF THE INVENTION**

This invention relates to a method of making a finished product and to the finished product so made.

It is well known to use wood and wood products to manufacture many different types of finished products. For example, wood fibres may be used to mould door skins, solid wood may be used to make mouldings, window frames, wall studding, decking, roof support rafters and bracing, plywood may be used for bulk handling bins, and the like. In addition, certain plastic materials may also be used to make structural items. For example, polyvinyl chloride may be used for the construction of sidings and window frames.

However, with the modern requirement for recyclability and the tendency for wood products to absorb water which results in thickness swell as well as microbial growth and a reduction in strength, there is a need for new products to replace such wood products.

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SUMMARY OF THE INVENTION

According to a first aspect of the invention there is provided a method of making a finished product including the steps of:

- (a) providing a feedstock comprising:
 - (1) 10 to 40 parts by weight of a polymer component comprising:
 - (i) 5 to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and
 - (ii) 0 to 95 parts by weight of polystyrene in particulate form;
 - (2) 10 to 70 parts by weight of a particulate extender; and
 - (3) 0 to 40 parts by weight of reinforcing fibres or particles;
- (b) mixing the feedstock with 10 to 35 parts by weight of a thermosetting resin selected from the group consisting of:
 - (i) an unsaturated polyester resin in liquid form and a catalyst for the polyester resin; and
 - (ii) an epoxy resin in liquid form and a catalyst for the epoxy resin; and
- (c) subjecting the product of step (b) to suitable conditions of temperature and pressure to cause the polyethylene and/or the polyvinyl chloride and the polystyrene, if present, to melt and the thermosetting resin to set to form the finished product.

According to a second aspect of the invention there is provided a finished product comprising a feedstock comprising:

- (1) 10 to 40 parts by weight of a polymer comprising :
 - (i) 5 to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in

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particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and

- (ii) 0 to 95 parts by weight of polystyrene in particulate form
 - (2) 10 to 70 parts by weight of a particulate extender; and
 - (3) 0 to 40 parts by weight of reinforcing fibres or particles;
- bound with 10 to 35 parts by weight of a thermosetting resin selected from the group consisting of:

- (iii) an unsaturated polyester resin in liquid form and a catalyst for the polyester resin; and
- (iv) an epoxy resin in liquid form and a catalyst for the epoxy resin.

Preferably, the feedstock comprises:

- (1) 15 to 35 parts by weight of the polymer component;
 - (2) 15 to 50 parts by weight of the extender;
 - (3) 0 to 25, preferably 10 to 25 parts by weight of the reinforcing fibres or particles;
- mixed with 15 to 25 parts by weight of the thermosetting resin.

It is to be noted that the finished product does not contain any hydraulic binder. In other words, the method of making the finished product is carried out without the use of a hydraulic binder and the final finished product does not include a hydraulic binder.

It is also to be noted that the method of the invention is carried out without the use of water at any stage.

The polymer component of feedstock may comprise polyethylene on its own, or polyvinyl chloride on its own, or a mixture of polyethylene and polyvinyl chloride without any other polymer, or any one of polyethylene, polyvinyl chloride or a mixture of polyethylene and polyvinyl chloride, together with an amount of polystyrene, all being in particulate form.

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The polymer component of the feedstock preferably comprises:

- (i) 40 to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and
- (ii) 0 to 60 parts by weight of polystyrene in particulate form.

The polymer component of the feedstock more preferably comprises:

- (i) at least 50 parts to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and
- (ii) 0 up to 50 parts by weight of polystyrene in particulate form.

The polyethylene and/or polyvinyl chloride provides the finished product with impact and flexural strength while the polystyrene provides the finished product with rigidity and hardness.

Depending upon the end use to which the finished product is to be put, the relative quantities of polyethylene, polyvinyl chloride and polystyrene may be varied to give a finished product with the desired properties.

The polyethylene may be chosen from linear low density polyethylene through to high density polyethylene.

The polyvinyl chloride may be any suitable polyvinyl chloride and is preferably a polyvinyl chloride that has been compounded with a stabiliser to prevent decomposition at the temperatures used in the method of the invention, and with a lubricant to propagate flow prior to and during the polymerisation of the thermosetting resin and to blend with the other components of the composition.

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The polystyrene is preferably milled polystyrene foam or polystyrene packaging.

The polyethylene, polyvinyl chloride and polystyrene must be in particulate form. By "particulate form" is meant round or flat particles, granules, and short fibres, all having a maximum dimension of 3 mm. The polyethylene, polyvinyl chloride and polystyrene are preferably used in powder form and preferably have a particle size of 1.0 mm diameter or less, more preferably 0.5 mm diameter or less.

The extender may be selected from the group consisting of a lightweight inorganic volume extender, a lightweight organic volume extender, and mixtures of two or more thereof.

The extender may also be milled mineral particles.

By a volume extender there is meant a particulate product with a low bulk density which when added to the composition of the invention increases its compression ratio, i.e the thickness of the product before pressing to the thickness of the product after pressing. The greater the compression ratio the greater is the control over product density, because the more the particles are pressed together, the better is the cohesion of the final product.

Volume extenders serve other useful functions including minimising lateral flow of the composition of the invention during compression moulding, as a function of its thermoplasticity.

In the composition of the invention, the use of volume extenders of different bulk densities and particle sizes allows for maximum flexibility in formulating a product for a particular application.

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The lightweight volume extender may be selected from hollow glass balloons, milled expanded perlite particles, undensified silica fume, exfoliated vermiculite particles, cork particles, leather particles, and a mixture of two or more thereof.

The extender, as indicated above, may also be milled mineral particles selected from the group consisting of calcium carbonate particles, silica particles, expanded clay particles, and the like.

The reinforcing fibres or particles may be selected from inorganic or organic fibres or particles.

As indicated above, the thermosetting resin may be selected from the group consisting of an unsaturated polyester resin and an epoxy resin.

The preferred thermosetting resin is an unsaturated polyester resin in liquid form with a catalyst for the polyester resin, for the reasons of controlled temperature of polymerisation initiation and minimal volatile products on polymerisation.

The extender is most preferably an inorganic extender, so as to avoid problems associated with the use of organic extenders.

The method of the invention may include a step between step (b) and step (c), of placing the product of step (b) on a first length of a sheet material or between first and second lengths of a sheet material so that in step (c) the first length and the second length, if present, are incorporated into the finished product.

Alternatively, the method of the invention may include a step after step (c) of:

- (a) placing the finished product on a first length of a sheet material or between first and second lengths of a sheet material with a layer of a

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thermosetting resin between the finished product and the first length and the second length, if present, and subjecting the resulting product to suitable conditions of temperature and pressure to laminate the first length to the finished product and to laminate the second length, if present, to the finished product, to form a composite product.

The sheet material may be for example a resin impregnated paper or a non-woven or woven fabric or the like, or a thermoplastic sheet material.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1A is a schematic view of a hydraulic press being used for the manufacture of a door skin of the invention;

Figure 1B is a side view of two door skins manufactured in the hydraulic press of Figure 1A;

Figure 2A is a schematic view of a hydraulic press being used for the manufacture of a moulding for window frames of the invention;

Figure 2B is a sectional side view of window frame mouldings manufactured in the hydraulic press of Figure 2A;

Figure 3 is a sectional side view of a wall stud of the invention;

Figure 4 is a sectional side view of decking of the invention;

Figure 5A is a sectional side view of a sheet which may be cut to form siding strips of the invention; and

Figure 5B is a sectional side view of an assembly of the siding strips of Figure 5A.

DESCRIPTION OF EMBODIMENTS

The crux of the invention is a method of making a finished product from a feedstock comprising a polymer component comprising polyethylene in particulate form and/or polyvinyl chloride in particulate form and optionally polystyrene in particulate form, an extender, and optionally reinforcing fibres or particles, by mixing the feedstock with a thermosetting resin and a catalyst for the resin. Thereafter the product is subjected to suitable conditions of temperature and pressure to cause the polyethylene and/or polyvinyl chloride and the polystyrene, if present, to melt and the thermosetting resin to set to form the finished product.

The first component is a feedstock comprising a polymer component comprising polyethylene and/or polyvinyl chloride and optionally polystyrene in particulate form.

The polyethylene may be for example a linear low density polyethylene for a finished product with toughness and flexibility or a high density polyethylene for a finished product with high rigidity.

The polyethylene may be either virgin polyethylene or, more preferably in terms of cost, be recycled or post industrial grade polyethylene, reduced to a relatively small particle size. When the polyethylene comes from a waste stream, it is acceptable for the waste stream and thus the polyethylene in the feedstock to contain a quantity of other polymers from the waste stream, such as, polyethylene terephthalate from bottles, polypropylene, polycarbonates, and polyesters. These additional waste polymers act as extenders for the feedstock in the finished product, as they generally have melting points higher than the temperatures used in step (c) of the method of the invention.

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For example, polyethylene terephthalate chips or fibres with a maximum dimension of 10 mm act advantageously as an extender in a finished product of the invention.

The polyethylene melts and commences to flow in the temperature range of from 115°C to 140°C inclusive.

The polyethylene must be in particulate, preferably powder form. Preferably, the polyethylene particles have a particle size of 1.0 mm in diameter or less, more preferably 0.5 mm in diameter or less, most preferably 150 microns in diameter or less.

The polyvinyl chloride may be for example a polyvinyl chloride that has been compounded with a stabiliser to prevent decomposition at the temperatures used in the method of the invention and with a lubricant to propagate flow prior to and during the polymerisation of the thermosetting resin and to blend with the other components of the composition. An example of a suitable polyvinyl chloride is DB178 by Polifin Division of Sasol, South Africa.

The properties of DB178 extrusion grade PVC are:

PROPERTIES	UNIT	TYPICAL VALUE	TEST METHOD
Relative Density		1.43	SABS Method 649
Shore D Hardness		83	ASTM D2240
Vicat Softening Point	°C	85	ASTM D1525
Tensile Strength	N/mm ²	38	BS2782 – 1970
Elongation	%	49	BS2782 – 1970
Volume Resistivity @ 23°C	Ωcm	28 x 10 ¹⁵	ASTM D257
Fire Rating	Self extinguishing		
Ash Content @ 650°C	%	4.30	

The polyvinyl chloride melts and commences to flow in the temperature range of from 150°C to 185°C.

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The polyvinyl chloride must be in particulate, preferably powder form. Preferably, the polyvinyl chloride particles have a particle size of 1.0 mm in diameter or less, more preferably 0.5 mm in diameter or less, most preferably 150 microns in diameter or less.

The polystyrene is preferably milled polystyrene foam or polystyrene packaging from a waste stream.

Polystyrene is a clear, glass-like material manufactured by the free radical polymerisation of phenylethene using benzoyl peroxide as an initiator. It has excellent thermal and electrical insulation properties and, in the method of the invention after having melted, it sets on cooling to a very hard inflexible, glass-like solid which is extremely resistant to water, having a water absorption after 24 hours of immersion of less than 0.06% by weight.

The polystyrene melts and commences to flow in the temperature range of from 100°C to 140°C inclusive.

The polystyrene must be in particulate, preferably powder form. Preferably, the polystyrene particles have a particle size of 1.0 mm in diameter or less, more preferably 0.5 mm in diameter or less, most preferably 150 microns in diameter or less.

The properties of polyethylene, polyvinyl chloride polystyrene and other polymers are set out in the table below.

Typical physical and mechanical comparisons between thermoplastics at 23°C are as follows:

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Material	Density (g/cm ³)	Degree of Crystallinity	Coefficient of Thermal Expansion Comparisons	Glass Transition Temp. (°C)
Polyethylene High-density	0.95-0.97	high	8.3-16.7	-120
Polyethylene Low-density	0.92-0.93	moderate	8.9-11.0	-120
Polypropylene	0.90-0.91	high	6.2	-20
Polystyrene	1.0-1.1	nil		100
Polyvinyl Chloride Unplasticized (PVC)	1.3-1.6	nil	2.8-3.3	85
Polytetrafluoroethylene	2.1-2.2	moderate to high		126

Material	Crystal Melting Temp (°C)	Tensile Strength (MPa)	Elongation at Break (%)	Flexural Modulus (GPa)
Polyethylene High-density	137	20-30	10-1000	1-15
Polyethylene Low-density	110	8-30	100-650	0.25-0.35
Polypropylene	176	30-40	100-600	1.2-1.7
Polystyrene	-	35-50	1-2	2.6-3.4
Polyvinyl Chloride Unplasticized (PVC)	-	40-50	2-80	2.1-3.4
Polytetrafluoroethylene	327	20-35	200-400	0.5

The feedstock also includes an extender.

The extender may be a lightweight inorganic volume extender such as hollow glass balloons, generally sourced from ground coal firing, which are siliceous, with a low bulk density in the range of from 200 g/l to 300 g/l and with a particle size in the range of from 50 to 300 microns, an example being Fillite SG; or milled expanded perlite particles such as Dicalite 411 or 471 by Chemserve Perlite South Africa, which has a similar bulk density to hollow glass micro balloons; or undensified silica fume with a bulk density in the range of 200 g/l to 300 g/l; or exfoliated vermiculite particles such as RSU by Palaborwa or MCF by Imerys, preferably with a particle size of less than 300 micron.

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The extender may also be a lightweight organic volume extender such as particles of cork or leather with bulk densities in the range of from 150 to 250 g/l inclusive.

The extender may also be milled mineral particles such as for example calcium carbonate, silica or expanded clay particles.

The extender may be a blend of two or more of the above.

The purpose of adding an extender to the feedstock is to control the density of the finished product with a minimum reduction in stiffness, and to minimise the coefficient of linear expansion in order to achieve acceptable dimensional stability, to improve stiffness and impact resistance, and to reduce cost.

The extender is preferably an inorganic extender selected from those set out above.

The most preferred extenders are selected from the group consisting of hollow glass balloons, expanded clay particles, exfoliated vermiculite particles and expanded perlite particles.

The feedstock may also comprise reinforcing inorganic particles or fibres such as glass fibre, either milled or up to 16 mm in length, but preferably of the order of 12 mm in length for best distribution and reinforcing; platelet minerals such as mica, preferably with a particle size in the range of 8 to 100 mesh, more preferably about 40 mesh, or phlogopite; or rod-like particles such as wollastonite.

The feedstock may also comprise reinforcing organic particles or fibres such as lignocellulosic fibres, e.g typha reed fibres, kenaf, flax, sisal and wood; or

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synthetic fibres such as polyester fibres capable of withstanding pressing temperatures in excess of 160°C.

The reinforcing fibres may be a blend of two or more of the above.

The next component is a thermosetting resin.

The first thermosetting resin which may be used is an unsaturated polyester resin in liquid form and a catalyst for the polyester resin.

An example of a suitable polyester resin is a high reactivity resin for hot pressed dough moulding compound applications (DMC), which resin is an orthophthalic, unsaturated polyester resin designed for use in the manufacture of glass and aggregate filled dough moulding compounds. It may be used in conjunction with low profile/low shrink additives for zero or low shrink hot press moulded products. Typical properties of this resin are a viscosity at 25°C of 1470 mPa.s, a volatile content of 35.5%, and a curing characteristic at 126°C using one part per hundred of Triganox 29B50 catalyst of a five minute gel time.

Another suitable resin is a chemical and water resistant isophthalic, neo-pentyl glycol unsaturated polyester resin used for high performance laminates. An example of this resin is NCS 993 by NCS Resins South Africa, which has a viscosity at 25°C of 540 to 800 mPa.s, an acid value of 10 to 16 mgKOH/g, a volatile content of 39 to 43%, which may be cured with a latent catalyst such as Triganox 29B50 which is a benzoyl peroxide.

Other suitable polyester resins include Crystic 272 and Crystic 196 from Scott Bader, England, catalysed by catalyst powder B; those sold under the Atlac brand from DSM; Polyite 33410, Polylite 8130, Polyite 8000 and Polyite 8382X from National Chemical Products, South Africa, catalysed with di-tert-

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butyl peroxy 3,3,5-tri-methylcyclohexane in dibutyl phthalate such as Triganox 29B240, Lucidol KL50, Triganox 21, Triganox C or Triganox K-70, benzoyl peroxide formulations, t-butyl perbenzoate from Interlox, e.g Codes TBTB and TBPB, dibenzoyl peroxide from Interlox Code BP.50-FT, and methyl isobutyl ketone peroxide from Interlox under Code MIKP NA1. These catalysts generally trigger polymerisation at a temperature in the region of 60°C upwards, more typically 80°C upwards.

The catalyst for the unsaturated polyester resin is preferably used in an amount of from 0.5% to 2.5% to 100% of the unsaturated polyester resin on a weight basis.

The second thermosetting resin which may be used is an epoxy resin in liquid form and a catalyst for the epoxy resin. A preferred epoxy resin is a low viscosity liquid epoxy resin manufactured from epichlorohydrin and bisphenol F. Typical examples of such resins include those manufactured by Shell Chemical Company under the names Epikote 816, 862, 232, 235 and 236, and those manufactured by Ciba-Geigy AG under the names XD.4150, XSA.214, Araldite AZ.15 and Araldite PY.340-2. Other suitable epoxy resins include epoxy resins with blocked amine functions, such as the reaction product of phthalic anhydride with diethylene triamine.

Suitable latent catalyst systems for use with epoxy resins include those catalyst systems supplied by Anchor Chemicals (UK) Limited such as Ancamine 2014S which is a modified polyamine; Anchor/catalyst 1786B which is 50/50 solution of p-toluene sulphonate of 2-amine-2-methyl-1-propanal in n-butanol.

These catalysts generally trigger polymerisation at a temperature of around 80°C or upwards.

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The unsaturated polyester resin may optionally be mixed with up to 50% by weight of the polyester resin of a styrene monomer for viscosity modification.

In order to propagate the adhesion between the polyethylene, polyvinyl chloride and polystyrene, i.e to induce the formation of a physical co-polymer between them and in order to impose upon the finished product improved toughness, and shock resistance when polystyrene is used in relatively large proportions, the product of the invention may include a thermoplastic elastomer, also known as a thermoplastic rubber or block co-polymer.

The thermoplastic elastomer is preferably used in an amount of from 2.5% to 35.0% based on 100% by weight of the polystyrene present.

The thermoplastic elastomer is preferably dissolved in a styrene monomer to produce a saturated solution. This saturated solution is then preferably blended with the thermosetting resin and its catalyst before this is added to the feedstock.

Examples of suitable thermoplastic elastomers are those having styrene end blocks and an elastomeric mid-block such as for example butadiene, isoprene, ethylene and the like, i.e those that have two different polymers in each molecule. Thus for example, suitable thermoplastic elastomers include a styrene/butadiene/styrene polymer, a styrene/isoprene polymer, and an acrylonitrile/butadiene/styrene polymer. The preferred thermoplastic elastomers are the styrene/butadiene polymers. Specific examples of suitable thermoplastic elastomers are the Kraton grades by Shell Chemicals. The "D" series are unsaturated and suitable for interior application and are comprised of styrene/isoprene/styrene block copolymers which are linear, and styrene/butadiene radial copolymers. The "G" series are fully hydrogenated grades for exterior applications and include styrene-ethylene/butylene-styrene block co-polymers which are linear and styrene-ethylene/propylene di-block

polymers. The Kraton "G" range of thermoplastic elastomers possess excellent resistance to oxygen, ozone and UV light degradation.

In addition, the propagation of adhesion between the thermosetting resin, polyethylene, polyvinyl chloride and polystyrene and any inorganic extender present may be further induced by the use of a silane coupling agent or crosslinker. An example is DC 1107 by Dow Corning used in an amount of about 0.5% on the weight of the thermosetting resin. This is a solvent soluble polymethylhydrogen siloxane. A further example is a gamma-methacryloxypropyltrimethoxy silane, Silquest A.174 silane by Witco Corporation which is specific to polyolefin to polyester to inorganic linkage, used in an amount of 0.1% by weight of the total composition.

In an alternative step, after step (c) the finished product may be placed on a first length of a sheet material or between first and second lengths of a sheet material, with a layer of a suitable thermosetting resin between the first length and the second length, if present, and the finished product, whereafter the whole is subjected to suitable conditions of temperature and pressure to laminate the first and second lengths of the sheet material to the finished product to form a composite product.

In step (b) of the method of the invention, the feedstock and the thermosetting resin are mixed.

It is to be noted that the finished product of the invention contains no hydraulic binder such as Portland cement.

In step (c) of the method of the invention, the product of step (b) is subjected to suitable conditions of temperature and pressure to cause the polyethylene and/or polyvinyl chloride and polystyrene, if present, to melt and the thermosetting resin to set to form the finished product.

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Suitable conditions of temperature and pressure include a temperature of from 110 to 200°C inclusive and a pressure of from 10 to 50 kg/cm² inclusive.

For example, the product of step (b) may be laid up between the platens of a press, and pressed to form a finished board product.

The exotherm from the thermosetting resin polymerization can elevate the composition temperature to above that of the platen temperature. Cooling of the product prior to releasing press platen contact is desirable to allow full strength and stability of the product to develop before handling.

Alternatively, the product of step (b) may be placed in a suitable mould and moulded to form a finished moulded product.

Further alternatively the product of step (b) may be extruded.

In a variation of the method of the invention, between step (b) and (c), the product of step (b) may be placed on a first length of a sheet material or between first and second lengths of a sheet material so that in step (c) the first length and the second length, if present, are incorporated into the finished product.

For example, the product of step (b) may be placed between first and second lengths of a sheet material, each length consisting of single or multi layers of a resin impregnated paper, preferably saturating Kraft paper impregnated with a resin selected from:

- 1 An unsaturated polyester resin with a catalyst therefor, optionally extended with a styrene monomer, in a solvent such as acetone. An example of a suitable unsaturated polyester resin is a neo-pentyl glycol unsaturated polyester resin, viz. NCS 993 by NCS Resins South Africa, in acetone, optionally including up to 10% by weight of a styrene

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monomer and also containing 0.5% to 2.5% by weight of the polyester resin of Triganox 29B50 catalyst.

- 2 A phenol formaldehyde resole resin such as Code J2018L by Borden Chemical Corporation, with an acid catalyst such as Phencat 10, (preferably in an amount of about 6% by weight of the resin) in methanol.
- 3 An MDI which is a diphenylmethane-4,4-diisocyanate, for example Suprasec 5005 or 2447 by Huntsman Corporation, optionally including a catalyst, in a suitable solvent such as acetone, ethyl acetate or dichloromethane.

The sheets of paper are impregnated with the resin system, whereafter the solvent is removed.

In step (c), the impregnated lengths of paper are then incorporated into the product of step (b) using suitable conditions of temperature and pressure as described above.

The Kraft paper preferably has a weight of about 200 to 400 g/m² and a thickness of approximately 350 to 500 microns.

Alternatively, and particularly where the finished product is to be shaped, for example to form a corrugated pallet deck, a non-woven fibrous sheet, preferably made of a polyester or polyethylene terephthalate, which both have melting points in excess of 245°C, may be used in place of the Kraft paper. These non-woven fibrous sheets may optionally be pre-impregnated with the same impregnating compositions described above for paper.

An example of a suitable non-woven fibrous material is Bidim Geotextile by Kaytech (Code A2 to Code A6) with a weight of 150 to 340 g/m², a tensile strength of 11 to 30 kNm, an elongation of 40% to 60% (which is necessary in

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pressing a shape to allow the accommodation of the increased surface area during compression moulding), and a melt temperature of about 250°C.

Other examples of a suitable sheet material are a chop strand matt glass fibre sheet of a mass of 300 to 600 g/m, sheet aluminium, and a melamine type high pressure laminate.

Again, the first and second lengths of a sheet material may be lengths of resin impregnated paper as described above.

The thermosetting resin for this purpose may be an unsaturated polyester resin, a phenol formaldehyde resole resin, or an MDI, preferably thickened with a suitable thickener such as Aerosil, or silica fume. The preferred thermosetting resin is an unsaturated polyester resin.

Once the composite product has been made as described above, further outer layers may be attached to the composite product. For example, on one or both sides of the composite product there may be attached an outer layer of a finished product of the invention.

Thus for example, there may be made a product comprising the following layers one on top of another:

an outer layer being a finished product of the invention formed from a linear low density polyethylene and an unsaturated polyester resin which has set;

an intermediate layer being a sheet of paper impregnated with a resin as described above;

a core being a finished product of the invention formed from a high density polyethylene and an unsaturated polyester resin which has set;

an intermediate layer of a sheet of paper impregnated with a resin as described above; and

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an outer layer being a finished product of the invention formed from a linear low density polyethylene and an unsaturated polyester resin which has set.

Any one of the layers may also include an amount of polystyrene.

Examples of the invention will now be given.

Example 1

A feedstock is prepared by mixing the following:

Irradiated high density polyethylene	700 parts by weight
Mica 40 mesh	200 parts by weight
Cenolite hollow glass balloons, 80 to 300 micron particle size	300 parts by weight

The feedstock is mixed with:

Isophthalic polyester resin, NCS 993 (NCS Resins)	400 parts by weight
Catalyst, Triganox 29B50	6 parts by weight
Styrene monomer	60 parts by weight

The mixture is pressed at a temperature of 160°C to a thickness of 11 mm at a density of 825 kg/m³ between first and second lengths of a sheet material, each of the first and second lengths comprising four sheets of resin impregnated Kraft paper with the weight of 200 g/m² per sheet.

The paper was impregnated with a composition comprising:

Isophthalic polyester resin, NCS 993 (NCS Resins)	20 parts by weight
Catalyst, Triganox 29B50	0.04 parts by weight
DC1107 cross-linking siloxane (Dow Corning)	0.20 parts by weight
Acetone	79 parts by weight
Accelerator, NCS ACI (NCS Resins) 0.06% of cobalt octoate in white spirit solution	0.10% parts by weight

The solvent is then removed.

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A layer of a thermosetting resin is located between the first and second lengths and the product, viz. NCS 993 isophthalic polyester resin (NCS Resins), catalysed with 1.5% Triganox 29B50 and thickened with Aerosil by Degussa. The result is a composite product with a thickness of 15 mm.

The composite product of the invention was tested under load in comparison to a 15 mm birch plywood.

The following test data for a sample with dimensions of 300 mm x 300 mm was obtained.

Maximum applied load – 806 kgf (7907N)

Deflection of board of the invention at maximum load 7.8 mm

Deflection of board of the invention at working load 4.45 mm

% strength compared to the 15 mm birch plywood based on the ultimate load 132%

% strength compared to the 15 mm birch plywood based working load deflection 106%

Example 2

An example of a composite suitable for siding or door skins is as follows:

	PERCENTAGE	WEIGHT UNIT
PVC DB178 (Sasol)	37	900
Fillite SG (Runcorn UK)	17	400
Mica 20 mesh	29	700
Orthophthalic Polyester Resin unsaturated Code 901 (NCS South Africa)	17	400
29B50 Triganox (Akzo Chemie)	-	4
DC 110 (Dow Corning)	-	4

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The board is pressed to a density of 1100kg/m^3 at a platen temperature of 185°C for 8 minutes to a thickness of 8 mm to produce a very strong composite of excellent dimensional stability.

Example 3

An example of a composition suitable for the production of high performance boards is as follows:

Component	Parts by weight
Polystyrene Packaging Waste Grade	150
Polyethylene High Density	700
Mica 40 Mesh	600
Hollow Glass Balloons	200
901 PA Resin Orthophthalic Unsaturated Polyester NCS	675
29B50 Triganox	8
Silane Cross Linker Dow Corning (Adhesion Promoter)	6
Glass Fibre 12mm length	1000
TOTAL	3339

The particle size of the polystyrene particles is 0.5 mm diameter and of the polyethylene is 80 mesh.

The above composition was mixed and then pressed at a pressure of 22 kg/cm^2 and at a temperature of 160°C for eight minutes to a thickness of 12 mm and a density of 1200 kg/cm^3 .

A board so produced was tested as a monolithic 12 mm board against a 12 mm 9 ply birch plywood board bonded with phenolic resins and surfaced with a B-stage phenolic resin over the placement sheet on both surfaces. The results are set out below.

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		Board of Invention	12mm 9 ply birch Shutter board
Property	Unit		
Tensile Strength	MPa	18.1	54.3
Tensile Modulus	MPa	534	1601
Flexural Strength	MPa	67.4	43.8
Flexural Modulus	MPa	2053	1063
Lap Shear	MPa	4.1	15.5
Water Absorption (Inc. 24 hrs)	%	0.176	30.89
Water Absorption (Inc. 7 days)	%	1.017	49.02
Area – (Inc. 24 hours)	%	0.100	1.477
Area – (Inc. 7 days)	%	0.139	1.929
Thickness – (Inc. 24 hrs)	%	0.353	3.658
Thickness – (Inc. 7 days)	%	0.441	6.983

Inc. 24 hrs means increase after 24 hours

The flexural strength and modulus of the board of the invention were respectively 1½ times and 2 times higher than that of the birch ply shutter board.

Various examples of finished products of the invention will now be described with reference to the accompanying drawings.

Referring to Figure 1A there is shown a platen 10 of a hydraulic press 12 for pressing a profiled shape. A mixture of the feedstock of the invention with an unsaturated polyester resin in liquid form and a catalyst for the polyester resin, is placed as a layer 14 on the platen 10. A profiled mould 16 is then used to press the mixture 14 to form a door skin moulding illustrated as 18 in Figure 1B.

As can be seen from Figure 1B, two door skin mouldings 18 may be joined to each other. A polyurethane foam may be injected into the space between them to product a polyethylene composite exterior door.

Referring to Figure 2A there is again shown a platen 20 of a hydraulic press 22 for pressing a profiled shape. A mixture of the feedstock, polyester resin and catalyst of the invention as described above is placed as a layer 24 on the platen 22. A profiled mould 26 is used to press the mixture 24 into the mould shapes 28. The mixture 24 first melts and then flows into the mould shapes 28 and then sets to form a moulding 30 illustrated in Figure 2B.

The moulding 30 may be cut into sections 32 for use as window frames.

The unsaturated polyester resin may be replaced by an epoxy resin to produce similar products:

Referring to Figure 3 there is shown a wall stud 40 made by the method of the invention which may be used in place of a typical 4 inch x 2 inch (10.16 cm x 5.08 cm) stud of timber. The stud 40 of the invention has a volume advantage over such a conventional timber stud of 25:1 but still has a density of 0.95 giving it adequate strength.

Referring to Figure 4 there is shown a sectioned side view of a length of decking 50, made by compression moulding according to the method of the invention.

Referring to Figure 5A there is shown a sectioned side view of a sheet 60 made by the method of the invention which may be cut to form siding strips 62.

Referring to Figure 5B, the siding strips 62 may be used in a conventional manner to provide a siding assembly.

Other finished products of the invention include corrugated roof sheeting, roof rafters and beams, dismountable bulk handling bins, pallets, stiles and rails for doors, window and door sills, shutter boards or form boards, and the like.

The finished products of the invention have the following advantages.

Polyethylenes, as used in extrusion or other processing, require to be cooled before solidification. The use of a polyester in the formulation of the invention allows the polyethylene first to flow, the catalyst then decomposes, and the polyester sets. The finished product so made has no memory, retains its shape, and may be demoulded at or near the process temperature.

By combining a thermoplastic material in dry powder form and a thermosetting resin in liquid form, there is no need for pre-compounding and thus much greater formulation flexibility can be achieved in terms of the quantity and type of extenders that may be included.

As the polyester resin used is reactive, as compared with polyethylene which is not reactive, the finished product of the invention may be painted or glued. This is not possible with an unmodified polyethylene product.

The finished products of the invention have considerable strength as a result of the materials used therein. In particular the thermosetting resin provides the strength needed for the heterogeneous nature of the thermoplastic resin(s) from a waste stream.

Polyesters and epoxies on polymerisation do not produce a gas, as do other resins such as phenol formaldehyde resole resins and isocyanate resins, which leads to processing advantages, such as the absence of porosity or blistering.

Because the mixture produced by blending the feedstock with the polyester or epoxy resin has the properties of both a thermoplastic and a thermoset material, in compression moulding this mixture may be induced to flow into shapes at a final density gradient of from 0.8 to 1.2, i.e 800 to 1200 kg/m³

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without materially diminishing the functionality of the finished product. Thus, far greater process flexibility can be achieved than would be possible by extrusion.

Polyethylene based extrusions or compression mouldings that contain extenders much beyond 20% by weight of the final product exhibit a dramatic loss of strength as a function of lack of compatibility or adhesion between the polyethylene and the extender. The use of a thermosetting resin in the finished product of the invention overcomes these problems.

The polyethylene and polystyrene used in manufacture of the finished product of the invention may both be derived from waste product streams, with cost advantages.

CLAIMS

1. A method of making a finished product includes the steps of:
 - (a) providing a feedstock comprising:
 - (1) 10 to 40 parts by weight of a polymer component comprising:
 - (i) 5 to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and
 - (ii) 0 to 95 parts by weight of polystyrene in particulate form;
 - (2) 10 to 70 parts by weight of a particulate extender; and
 - (3) 0 to 40 parts by weight of reinforcing fibres or particles;
 - (b) mixing the feedstock with 10 to 35 parts by weight of a thermosetting resin selected from the group consisting of:
 - (i) an unsaturated polyester resin in liquid form and a catalyst for the polyester resin; and
 - (ii) an epoxy resin in liquid form and a catalyst for the epoxy resin; and
 - (c) subjecting the product of step (b) to suitable conditions of temperature and pressure to cause the polyethylene and/or the polyvinyl chloride and the polystyrene, if present, to melt and the thermosetting resin to set to form the finished product.
2. A method according to claim 1 which comprises:
 - (a) a feedstock comprising:
 - (1) 15 to 35 parts by weight of the polymer component;
 - (2) 15 to 50 parts by weight of the extender;
 - (3) 0 to 25 parts by weight of the reinforcing fibres or particles;
 - (b) mixed with 15 to 25 parts by weight of the thermosetting resin.

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3. A method according to claim 1 or claim 2 wherein the polymer component of the feedstock comprises:
 - (i) 40 to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and
 - (ii) 0 to 60 parts by weight of polystyrene in particulate form.
4. A method according to claim 3 wherein the polymer component of the feedstock comprises:
 - (i) at least 50 parts to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and
 - (iii) 0 up to 50 parts by weight of polystyrene in particulate form.
5. A method according to any one of claims 1 to 4 wherein the polyethylene, the polyvinyl chloride, and the polystyrene are used in powder form having a particle size of 1.0 mm diameter or less.
6. A method according to any one of claims 1 to 5 wherein the thermosetting resin is an unsaturated polyester resin in liquid form and a catalyst for the polyester resin.
7. A method according to any one of claims 1 to 6 wherein the extender is selected from the group consisting of a lightweight inorganic volume extender, a lightweight organic volume extender, milled mineral particles, and mixtures of two or more thereof.

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8. A method according to claim 7 wherein the extender is selected from the group consisting of hollow glass balloons, milled expanded perlite particles, undensified silica fume, exfoliated vermiculite particles, cork particles, leather particles, calcium carbonate particles, expanded clay particles, silica particles, and a mixture of two or more thereof.
9. A method according to any one of claims 1 to 8 wherein the reinforcing fibres or particles are selected from the group consisting of glass fibres, mica particles, phlogopite particles, wollastonite particles, lignocellulosic fibres, synthetic fibres, and a mixture of two or more thereof.
10. A method according to any one of claims 1 to 9 wherein in step (c) the product of step (b) is subjected to a temperature of from 110°C to 200°C inclusive and a pressure of from 10 kg/cm² to 50 kg/cm² inclusive.
11. A method according to any one of claims 1 to 10 wherein the method includes a step between step (b) and step (c) of placing the product of step (b) on a first length of a sheet material or between first and second lengths of a sheet material so that in step (c) the first length and the second length, if present are incorporated into the finished product.
12. A method according to any one of claims 1 to 10 wherein the method includes a step after step (c) of:
 - (a) placing the finished product on a first length of a sheet material or between first and second lengths of a sheet material with a layer of a thermosetting resin between the finished product and the first length and the second length, if present, and subjecting the resulting product to suitable conditions of temperature and pressure to laminate the first length to the finished product and to laminate the second length, if present, to the finished product, to form a composite product.

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13. A method according to claim 11 or claim 12 wherein the sheet material is selected from the group consisting of a resin impregnated paper, a non-woven fabric, a woven fabric and a thermoplastic sheet material.
14. A finished product comprises a feedstock comprising:
 - (1) 10 to 40 parts by weight of a polymer component comprising:
 - (i) 5 to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and
 - (ii) 0 to 95 parts by weight of polystyrene in particulate form;
 - (2) 10 to 70 parts by weight of a particulate extender; and
 - (3) 0 to 40 parts by weight of reinforcing fibres or particles bound with 10 to 35 parts by weight of a thermosetting resin selected from the group consisting of:
 - (iii) an unsaturated polyester resin in liquid form and a catalyst for the polyester resin; and
 - (iv) an epoxy resin in liquid form and a catalyst for the epoxy resin.
15. A finished product according to claim 14 which comprises:
 - (1) 15 to 35 parts by weight of the polymer component;
 - (2) 15 to 50 parts by weight of the extender;
 - (3) 0 to 25 parts by weight of the reinforcing fibres or particles; bound with 15 to 25 parts by weight of the thermosetting resin.
16. A finished product according to claim 14 or claim 15 wherein the polymer component of the feedstock comprises:
 - (i) 40 to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and

- (ii) 0 to 60 parts by weight of polystyrene in particulate form.
17. A finished product according to claim 16 wherein the polymer component of the feedstock comprises:
- (i) at least 50 parts to 100 parts by weight of a polymer selected from the group consisting of polyethylene in particulate form, polyvinyl chloride in particulate form, and a mixture of polyethylene and polyvinyl chloride both in particulate form; and
 - (ii) 0 up to 50 parts by weight of polystyrene in particulate form.
18. A finished product according to any one of claims 14 to 17 wherein the polyethylene, the polyvinyl chloride, and the polystyrene are used in powder form having a particle size of 1.0 mm diameter or less.
19. A finished product according to any one of claims 14 to 18 wherein the thermosetting resin is an unsaturated polyester resin in liquid form and a catalyst for the polyester resin.
20. A finished product according to any one of claims 14 to 19 wherein the extender is selected from the group consisting of a lightweight inorganic volume extender, a lightweight organic volume extender, milled mineral particles, and mixtures of two or more thereof.
21. A finished product according to claim 20 wherein the extender is selected from the group consisting of hollow glass balloons, milled expanded perlite particles, undensified silica fume, exfoliated vermiculite particles, cork particles, leather particles, calcium carbonate particles, expanded clay particles, silica particles, and a mixture of two or more thereof.

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22. A finished product according to any one of claims 14 to 21 wherein the reinforcing fibres are selected from the group consisting of glass fibres, mica particles, phlogopite particles, wollastonite particles, lignocellulosic fibres, synthetic fibres, and a mixture of two or more thereof.

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Fig 1A

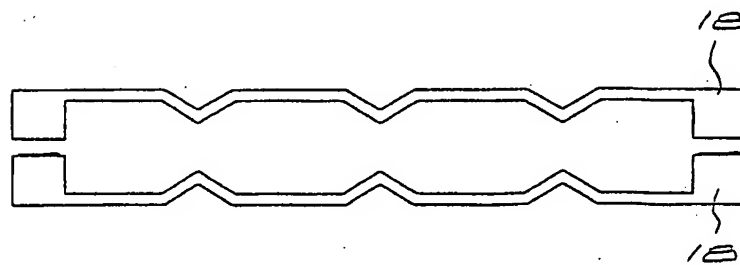
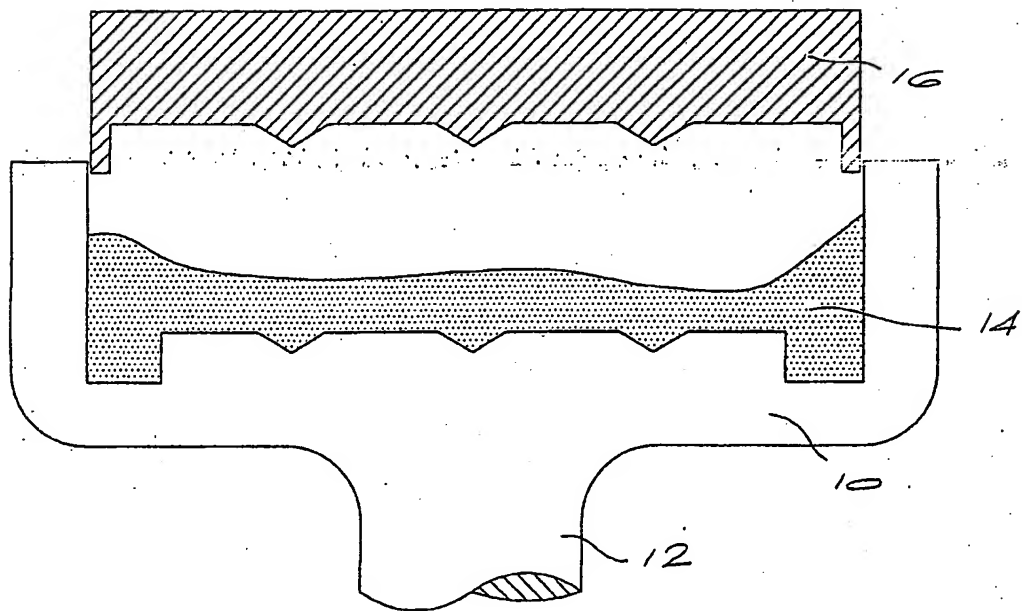
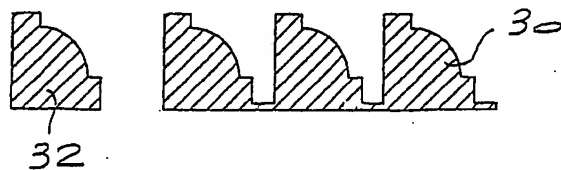
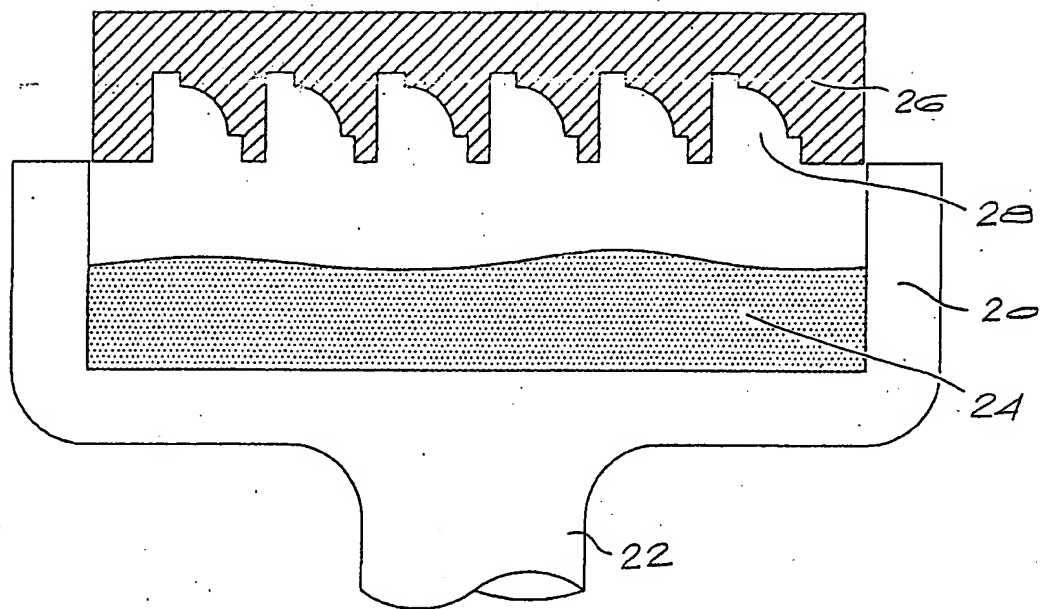


Fig 1B

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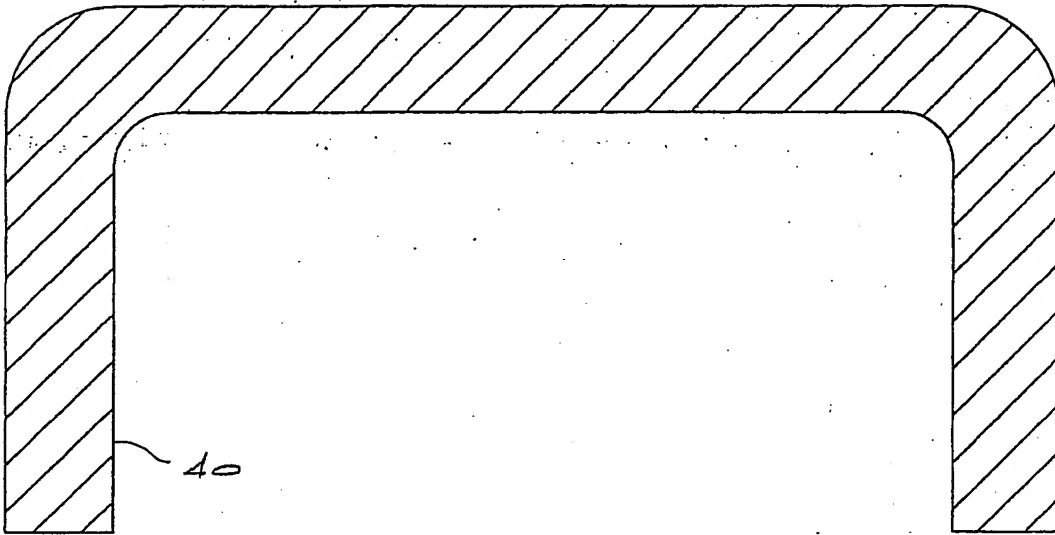
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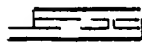


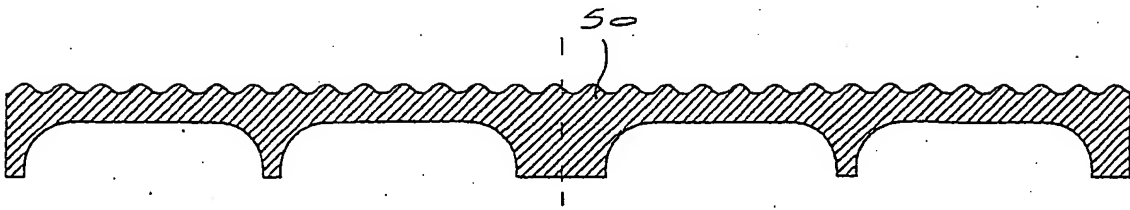
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3/4

 3

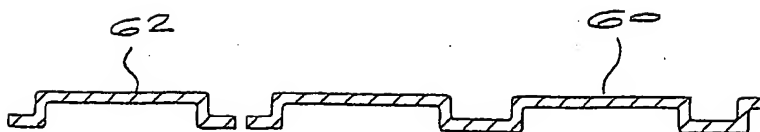
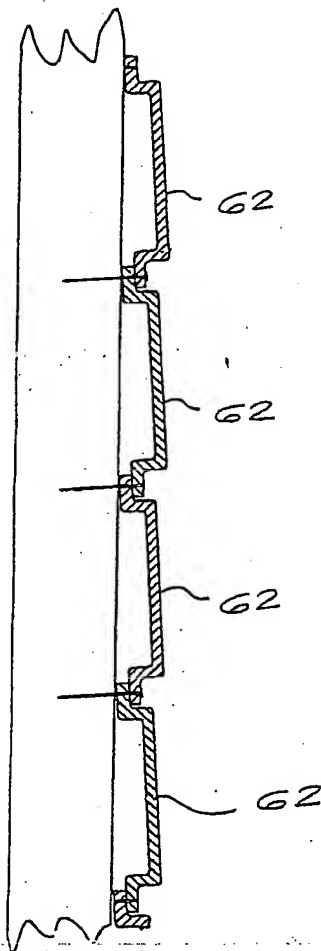


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5B



5A

International Application No
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IPC 7 C08J5/00 C08L27/06 C08L23/06 C08J5/04

B. FIELDS SEARCHED

IPC 7 C08J C08L

Electronic data base consulted during the International search (name of data base and, where practical, search terms used)

WPI Data, PAJ, EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 786 004 A (ADACHI T ET AL) 15 January 1974 (1974-01-15) column 14, line 7,21,43,63-70 example 8 claims 1,11 ----	1-22
X	US 5 895 790 A (GOOD ELMER) 20 April 1999 (1999-04-20) claims 1-3 column 12, line 24-48 ----	1-22
X	US 5 932 641 A (TROUVE PATRICK ET AL) 3 August 1999 (1999-08-03) column 4, line 1-8 claims 1-4 ----- -/-	1-22

☒ Further documents are listed in the continuation of box C.

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Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax (+31-70) 340-3016

Authorized officer

Natus, G

INTERNATIONAL SEARCH REPORT

WO 03/048238

International Application No
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 331 507 A (ACR CO LTD ;ASAHI DENKA KOGYO KK (JP)) 6 September 1989 (1989-09-06) page 11, line 50-65 claim 1 ----	1-22
X	EP 0 747 444 A (OTSUKA KAGAKU KK) 11 December 1996 (1996-12-11) claims 1-8 ----	1-22
X	GB 936 351 A (BRITISH INDUSTRIAL PLASTICS) 11 September 1963 (1963-09-11) claim 1 ----	1-22
X	FR 2 682 117 A (MENZOLIT) 9 April 1993 (1993-04-09) claim 1 page 5, line 14 ----	1-22
X	US 4 727 096 A (CHOUDIN CLAUDE) 23 February 1988 (1988-02-23) claim 1 -----	1-22

INTERNATIONAL SEARCH REPORT

WO 03/048738

International Application No

PCT/IB PCT/IB02/05100

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 3786004	A	15-01-1974	JP 50025518 B	23-08-1975
			JP 49002331 B	19-01-1974
			CA 972900 A1	12-08-1975
			DE 2155655 A1	20-07-1972
			FR 2120700 A5	18-08-1972
			GB 1373766 A	13-11-1974
			IT 944216 B	20-04-1973
US 5895790	A	20-04-1999	NONE	
US 5932641	A	03-08-1999	FR 2722204 A1	12-01-1996
			CA 2153358 A1	09-01-1996
			EP 0691336 A1	10-01-1996
EP 0331507	A	06-09-1989	JP 1225650 A	08-09-1989
			JP 2763540 B2	11-06-1998
			JP 1252650 A	09-10-1989
			JP 2632907 B2	23-07-1997
			JP 2073842 A	13-03-1990
			JP 2693788 B2	24-12-1997
			AT 89582 T	15-06-1993
			DE 68906569 D1	24-06-1993
			DE 68906569 T2	09-09-1993
			EP 0331507 A1	06-09-1989
			ES 2057107 T3	16-10-1994
			US 4977201 A	11-12-1990
EP 0747444	A	11-12-1996	EP 0747444 A1	11-12-1996
			KR 233359 B1	01-12-1999
			US 5952416 A	14-09-1999
			WO 9618687 A1	20-06-1996
			JP 8231859 A	10-09-1996
GB 936351	A	11-09-1963	DE 1192820 B	13-05-1965
			FR 1291844 A	27-04-1962
			NL 120820 C	
			NL 264053 A	
FR 2682117	A	09-04-1993	FR 2682117 A1	09-04-1993
			AT 152088 T	15-05-1997
			DE 69219349 D1	28-05-1997
			DE 69219349 T2	13-11-1997
			EP 0607330 A1	27-07-1994
			ES 2103383 T3	16-09-1997
			WO 9307102 A1	15-04-1993
US 4727096	A	23-02-1988	FR 2589473 A1	07-05-1987
			AT 56032 T	15-09-1990
			BR 8605337 A	04-08-1987
			CA 1276339 A1	13-11-1990
			DE 3673772 D1	04-10-1990
			DK 517186 A	01-05-1987
			EP 0225234 A1	10-06-1987
			JP 62106958 A	18-05-1987
			NO 864318 A ,B,	04-05-1987
			NO 910809 A ,B,	04-05-1987
			PT 83639 A ,B	01-11-1986

Original Application No

A. CLASSIFICATION OF SUBJECT MATTER

According to International Patent Classification (IPC) or to both national classification and IPC

Minimum documentation searched (classification system followed by classification symbols)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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Date of the actual completion of the international search

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Name and mailing address of the ISA
European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Natus, G

INTERNATIONAL SEARCH REPORT

I International Application No

PCT/JP 02/05100

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X	EP 0 331 507 A (ACR CO LTD ;ASAHI DENKA KOGYO KK (JP)) 6 September 1989 (1989-09-06) page 11, line 50-65 claim 1 ---	1-22
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X	GB 936 351 A (BRITISH INDUSTRIAL PLASTICS) 11 September 1963 (1963-09-11) claim 1 ---	1-22
X	FR 2 682 117 A (MENZOLIT) 9 April 1993 (1993-04-09) claim 1 page 5, line 14 ---	1-22
X	US 4 727 096 A (CHOUDIN CLAUDE) 23 February 1988 (1988-02-23) claim 1 -----	1-22

INTERNATIONAL SEARCH REPORT

onal Application No

PCT/IB 02/05100

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 3786004	A	15-01-1974	JP 50025518 B	23-08-1975
			JP 49002331 B	19-01-1974
			CA 972900 A1	12-08-1975
			DE 2155655 A1	20-07-1972
			FR 2120700 A5	18-08-1972
			GB 1373766 A	13-11-1974
			IT 944216 B	20-04-1973
US 5895790	A	20-04-1999	NONE	
US 5932641	A	03-08-1999	FR 2722204 A1	12-01-1996
			CA 2153358 A1	09-01-1996
			EP 0691336 A1	10-01-1996
EP 0331507	A	06-09-1989	JP 1225650 A	08-09-1989
			JP 2763540 B2	11-06-1998
			JP 1252650 A	09-10-1989
			JP 2632907 B2	23-07-1997
			JP 2073842 A	13-03-1990
			JP 2693788 B2	24-12-1997
			AT 89582 T	15-06-1993
			DE 68906569 D1	24-06-1993
			DE 68906569 T2	09-09-1993
			EP 0331507 A1	06-09-1989
			ES 2057107 T3	16-10-1994
			US 4977201 A	11-12-1990
EP 0747444	A	11-12-1996	EP 0747444 A1	11-12-1996
			KR 233359 B1	01-12-1999
			US 5952416 A	14-09-1999
			WO 9618687 A1	20-06-1996
			JP 8231859 A	10-09-1996
GB 936351	A	11-09-1963	DE 1192820 B	13-05-1965
			FR 1291844 A	27-04-1962
			NL 120820 C	
			NL 264053 A	
FR 2682117	A	09-04-1993	FR 2682117 A1	09-04-1993
			AT 152088 T	15-05-1997
			DE 69219349 D1	28-05-1997
			DE 69219349 T2	13-11-1997
			EP 0607330 A1	27-07-1994
			ES 2103383 T3	16-09-1997
			WO 9307102 A1	15-04-1993
US 4727096	A	23-02-1988	FR 2589473 A1	07-05-1987
			AT 56032 T	15-09-1990
			BR 8605337 A	04-08-1987
			CA 1276339 A1	13-11-1990
			DE 3673772 D1	04-10-1990
			DK 517186 A	01-05-1987
			EP 0225234 A1	10-06-1987
			JP 62106958 A	18-05-1987
			NO 864318 A ,B,	04-05-1987
			NO 910809 A ,B,	04-05-1987
			PT 83639 A ,B	01-11-1986

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SECRET

DECLASSIFIED